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Published in:
Applied Physics Letters

Link to article, DOI:
[10.1063/1.5009168](https://doi.org/10.1063/1.5009168)

Publication date:
2018

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Pedrinazzi, P., Caridad, J. M., Mackenzie, D. M. A., Pizzocchero, F., Gammelgaard, L., Jessen, B. S., Sordan, R., Booth, T. J., & Bøggild, P. (2018). High-quality graphene flakes exfoliated on a flat hydrophobic polymer. *Applied Physics Letters*, 112(3), [033101]. <https://doi.org/10.1063/1.5009168>

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Supplementary Material

High-quality graphene flakes exfoliated on a flat hydrophobic polymer

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1. Optical image of graphene on PMMA/PVA

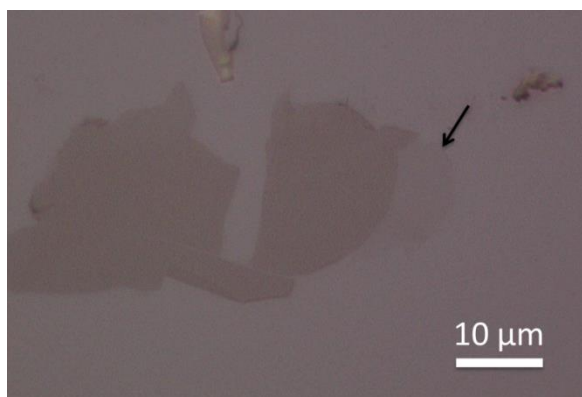


Figure S1. Optical image of a graphene flake on PMMA/PVA (black arrow).

2. Extraction of quantitative values of strain and doping in high-quality graphene flakes on PMMA/PVA

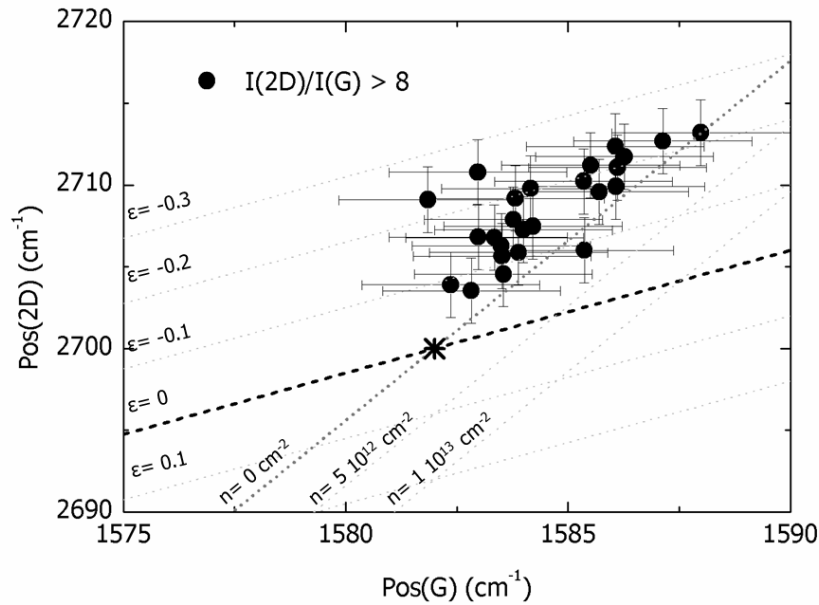


Figure S2. Strain and doping analysis of PMMA-supported single layer graphene with ratios $I(2D)/I(G) > 8$ via Raman quantities of Pos(2D) and Pos(G). The predicted evolution in the unique presence of strain (grey dotted line) or doping (black dashed line) are also plotted, and intersect at a point (asterisk) representing doping- and strain-free graphene ($Pos(G)^0$, $Pos(2D)^0$). Thinner lines parallel to the strain line are separated by a difference in doping levels of $5 \cdot 10^{12} \text{ cm}^{-2}$. Thinner lines parallel to the doping line are separated by a difference in strain levels of 0.1%. Within our experimental error ($\pm 2 \text{ cm}^{-1}$), we extract averaged strain levels of -0.17% and common doping levels in our flakes below $2.5 \cdot 10^{12} \text{ cm}^{-2}$.

3. Raman spectra of graphene flakes on PMMA/PVA and after hBN encapsulation

We measured the evolution of the Raman spectra of graphene flakes initially exfoliated on PMMA/PVA and subsequently encapsulated between hBN. Showing how the high $I(2D)/I(G)$ ratios are still observed when changing the graphene environment (substrate), we can rule out the possibility that the high $I(2D)/I(G)$ ratios appearing on PMMA/PVA are merely due to convenient interference effects arising from selected dielectric layers^{S1}.

Table S1 shows the averaged Raman parameters of two graphene flakes before and after the encapsulation process. $I(2D)/I(G)$ ratios are high in both cases, showing differences between pre- and post-encapsulation values within 25%. We attribute this variation to the still complex interplay between residual strain and contaminants occurring when performing the encapsulation process in air, an issue which could be improved by atmospheric control during the encapsulation step^{S2-S4}.

	$I(2D)/I(G)$	FWHM(G) (cm^{-1})	FWHM(2D) (cm^{-1})
Flake 1 (on PMMA/PVA)	8.2	14	22
Flake 1 (encapsulated)	9.1	13.5	20.5
Flake 2 (on PMMA/PVA)	8.5	13.5	21
Flake 2 (encapsulated)	7.3	12	21

Table S1. Averaged Raman parameters (over 5 random points) of two graphene flakes before and after the encapsulation process.

Furthermore, we show Raman $I(2D)/I(G)$ maps of the full encapsulated graphene flakes in order to demonstrate the homogeneity of the obtained stacks. The $I(2D)/I(G)$ map of encapsulated Flake 1 is shown in Figure 3 (main text). Meanwhile, Raman map of the $I(2D)/I(G)$ ratio of the encapsulated Flake 2 is shown in Figure S3. We remark the high homogeneity existent even in larger ($\sim 200\mu\text{m}^2$) flakes such as Flake 2. Existing inhomogeneities in the periphery of the flake are due to a multilayer part or bent areas (see optical contrast image).

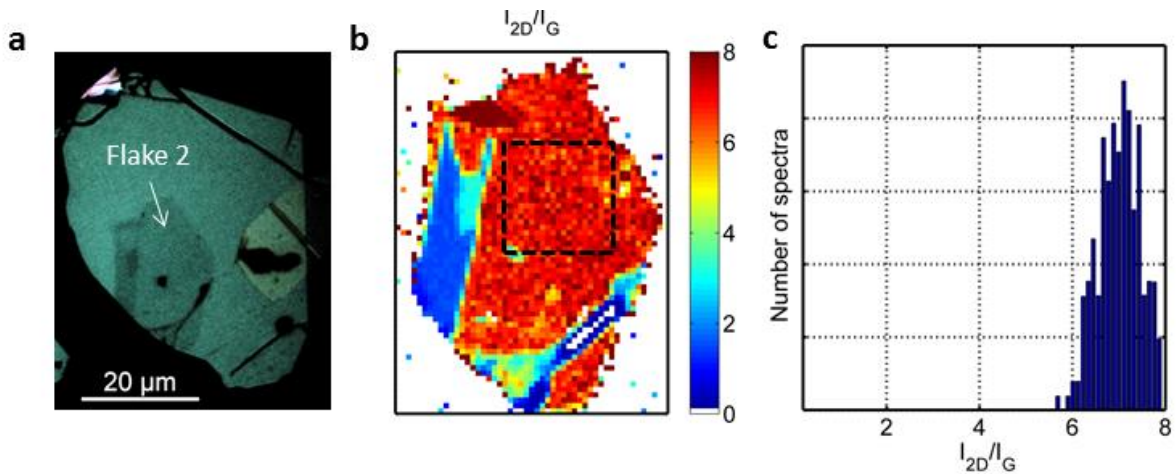


Figure S3. (a) Optical image (contrast) of the encapsulated Flake 2. (b) Map of the $I(2D)/I(G)$ ratio of

encapsulated Flake 2. (c) Histogram of the $I(2D)/I(G)$ values in the middle of Flake 2 (dashed square in (b)), where zones with multilayer graphene close to the edges are excluded.

4. Device homogeneity

The homogeneity of the fabricated graphene devices was investigated by comparing the resistance R in two perpendicular configurations^{S5} (A and B, rotated by 90 degrees). Figure S4 shows both measurements for the device fabricated from Flake 1 (Figure 3, main text). Resistance values are very close, almost overlapping except for gate-voltage (V_g) values close to the charge neutrality point. The latter can be due to residual charge inhomogeneities^{S5,S6} still present on the graphene surface or at its edges.

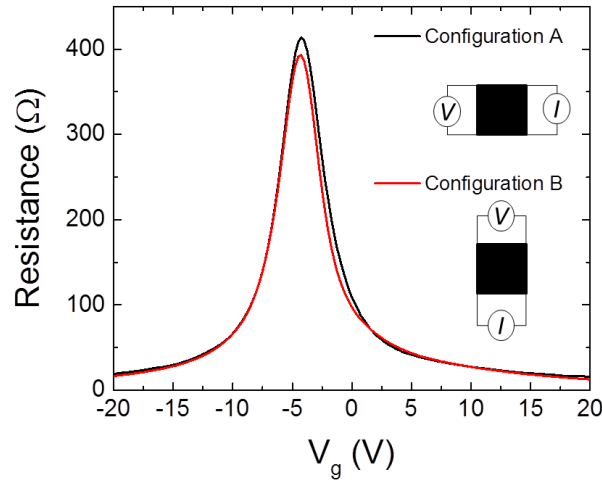


Figure S4. Four point resistance measurements ($R=V/I$) of a 5×5 micrometer square device made from Flake 1 in two perpendicular configurations (A and B).

5. Graphene on PMMA/SiO₂ substrates

We show that samples with high $I(2D)/I(G)$ ratio can be obtained on PMMA/SiO₂ substrates. We remark that these substrates are different of those from Fig.1 and 2 in the main text, where graphene was exfoliated on PMMA/PVA/SiO₂ substrates. By varying the substrate composition and thickness, we exclude that interference effects would be the cause of the high $I(2D)/I(G)$ ratio

achieved in our graphene flakes . As shown in Fig. S5, flakes with high ratios (>4) are obtained in this case, too.

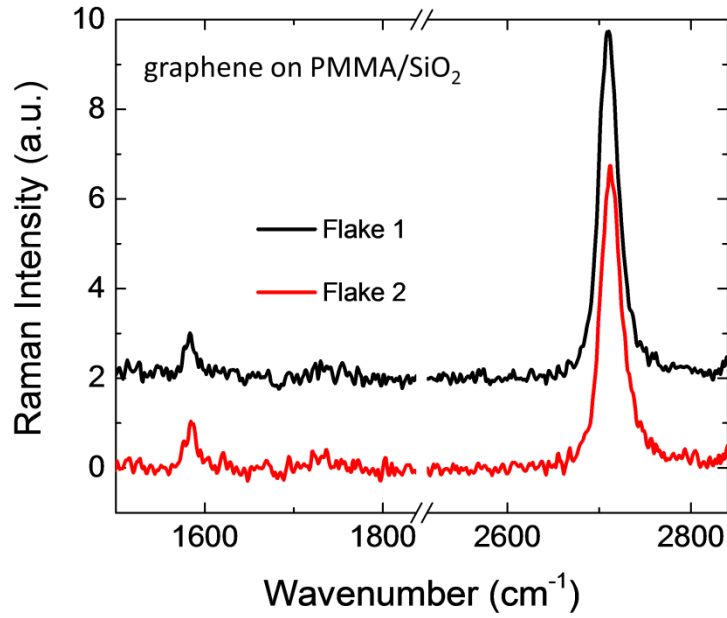


Figure S5. Raman spectra of two graphene flakes exfoliated on PMMA/SiO₂ substrates showing ratios of ~ 8 and ~ 6 , respectively.

6. Surface roughness of spin-coated PVA films

For completeness, we show the surface roughness of spin-coated PVA films. These films are spin-coated between SiO₂ and PMMA in the majority of our exfoliated flakes.

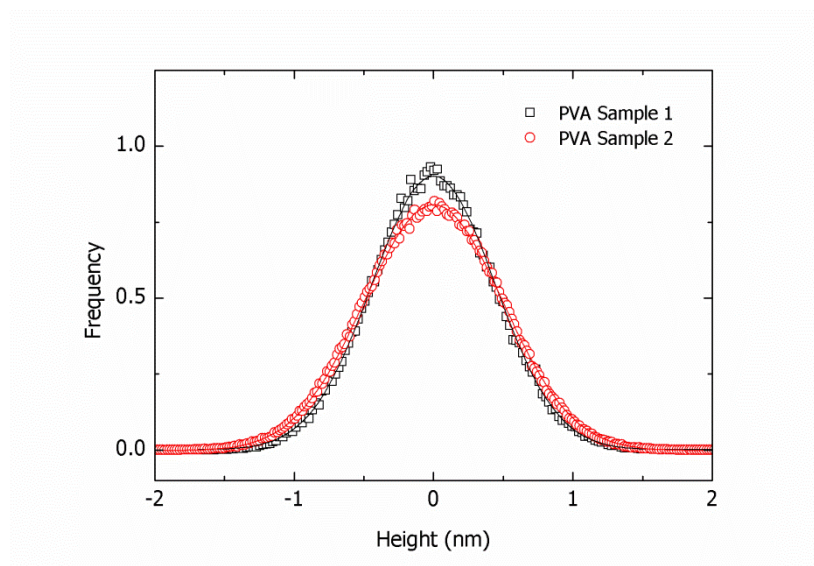


Figure S6. Characterization of the surface roughness of PVA films via AFM. The figure shows histogram of the height distribution (surface roughness) measured for two PVA samples on $1\mu\text{m}^2$ windows. The measured surface roughness of this polymer (given by the standard deviation of a fitted Gaussian) is >0.35 nm, much larger (>0.34 nm) than the one obtained for our PMMA layers (~ 0.16 nm) and even SiO_2 substrates (~ 0.22 nm), see Figure 1b.

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